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FINAL REPORT ON DEVELOPMENT & EVALUATION OF TASK GROUP 3 PRESSURE TUBES

Report No 83-457-K

By

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ABSTRACT

This report describes the production and evaluation of pressure tubes manufactured to the recommendations of Task Group 3 (TG3) of the Creep Engineering Design Plan. The Zr-2.5 wt% Nb tubes were manufactured by modified production routes to change their metallurgical structure and so reduce the in-service elongation rates. Three modified routes were investigated and a total of twenty-eight tubes produced. There were no difficulties in manufacture and the tubes satisfied the quality assurance and design specifications of reactor grade tubes. Metallurgical evaluation showed that the expected changes in microstructure had occurred but not to the extent anticipated. The TG3 tubes were found to have comparable properties to current tubes when tested for: tensile strength (irradiated and unirradiated); hydride cracking; stress to reorient hydrides; hydrogen diffusion; flaw tolerance; corrosion (irradiated and unirradiated); wear; rolled joint characteristics; irradiation creep and growth. Lower in-service elongation rates are expected for tubes produced by two of the modified routes.

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EXECUTIVE SUMMARY

FINAL REPORT ON DEVELOPMENT & EVALUATION OF TASK GROUP 3 PRESSURE TUBES

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To investigate the impact of in-service deformation of core components on reactor operation, Ontario Hydro formed in 1978 the Creep Engineering Design Plan. This report describes the development and evaluation of Zr-2.5 wt% Nb pressure tubes as recommended by Task Group 3 of the CEDP. TG3 was concerned with the production of tubes with lower in-service elongation rates than current tubes. Three different post extrusion production routes were used and a total of twenty-eight tubes manufactured. There were no difficulties encountered in manufacture and the tubes satisfied the quality assurance and design specifications of reactor grade tubes. Metallurgical evaluation of the tubes compared their properties with current tubes. The properties investigated were: tensile strength (irradiated and unirradiated); hydride cracking; stress to re-orient hydrides; hydrogen diffusion; flaw tolerance; corrosion; wear; rolled joints; microstructure and irradiation creep and growth. In all respects the TG3 tubes compare favourably with current tubes. The irradiation growth results indicate that tubes produced by two of the routes would have lower elongation rates than current tubes.

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B	Costs of Development Program
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D	Development and Hold Point Evaluation
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To Mr. F.J. Kee
Director of Research

FINAL REPORT ON DEVELOPMENT & EVALUATION
OF TASK GROUP 3 PRESSURE TUBES

1.0 INTRODUCTION

Pressure tubes for CANDU reactors have been manufactured since 1957 by a process of extrusion and cold drawing. The development followed closely the fabrication of the first zirconium alloy pressure tubes for the PRTR, CVTR and N-reactors in the United States which had been produced by a number of manufacturers using different fabrication routes/1/. Early CANDU reactors - NPD, Douglas Point and Pickering 1 and 2, used Zircaloy-2. The Zr-2.5 wt% Nb (Zr-2.5 Nb) alloy was selected for Pickering 3 and 4 because its higher strength permitted the use of a thinner wall tube with a resultant advantage in neutron economy over Zircaloy-2 and it has been used in all subsequent CANDU reactors.

The extrusion and cold drawing fabrication process developed corrosion resistance and creep resistance and adequate uniform tensile properties. Product development until recently has been associated with improving the dimensional, surface and defect quality of the tubes and the uniformity of structure and properties. Supporting research has been toward understanding the relationship between fabrication parameters and final properties.

Measurements of pressure tube elongation early in the life of Pickering Generating Station 'A' (PGSA) revealed that the elongation rates were much higher than had been anticipated in the design of PGSA. To investigate the impact of in-service deformation of core components on the operation of reactors Ontario Hydro formed, in 1978, the Creep Engineering Design Plan (CEDP). The work of the CEDP was performed by three Task Groups which involved personnel from AECL and various Ontario Hydro Divisions.

Task Group 3 (TG3) concentrated on the production of pressure tubes which would have lower elongation rates than current tubes. TG3 also considered the use of alternative alloys for the manufacture of pressure tubes and modifications to calandria tube fabrication to reduce the rate of fuel channel sag and calandria tube elongation. This report is concerned with the development of alternative production routes and the manufacture by these

routes of Zr-2.5 Nb pressure tubes. The development of alternative alloys is an ongoing project at AECL-RC (Chalk River) and the calandria tube studies are on a relatively small scale and will be covered in a separate report. For detailed information on the recommendations of Task Group 3, see Appendix A. (The Appendices to this report are issued as separate documents).

Pressure tube elongation is considered to occur as a result of irradiation enhanced creep and growth. Task Group 3 evaluated the available information on how metallurgical structure influenced irradiation creep and growth. The major source of information was from studies/1,2,3/ on Zircaloy pressure tubes manufactured by various fabrication routes which resulted in differences in metallurgical structure (texture, dislocation density and grain shape). Basically the information on Zircaloy was applied to Zr-2.5 Nb pressure tubes. The extent of the changes to the fabrication route were restricted by the capabilities of the current manufacturer. Because the quantitative effects of each fabrication parameter on the metallurgical structure of Zr-2.5 Nb were not known, three production routes were proposed (Figure 1). Laboratory simulation of the proposed routes indicated that adequate tensile strength would be achieved (Appendix D).

This report gives an overview of the various aspects of the pressure tube development program as recommended by Task Group 3 of the Creep Engineering Design Plan. The details of the various developmental, fabrication and evaluation studies are contained in the Appendices which are issued as separate documents from this report.

2.0 FABRICATION

The fabrication exercise was characterized by a number of features:

a) The tubes were procured using the quality assurance practice for power reactor tubes. Thus the procedure of each fabrication operation was reviewed and approved by standard procurement engineering - quality assurance practice against specification requirements. This provided a basis for any subsequent potential recommendation to install any of the tubes in a power reactor. The manufacturer was required to produce detailed history dockets at the completion of fabrication.

b) The routes were selected so that tubes could be made using existing equipment and technology.

c) There were several hold points at which approval to proceed was based on testing the material to simulate subsequent treatment.

d) The planning and selection of fabrication steps was done after detailed consultation with three manufacturers.

Teledyne Wah Chang Albany melted the ingot and reduced it to the machined billet condition. Chase Nuclear extruded the billets and cold-worked the extrusions to final size. Bristol Aerospace carried out the intermediate vacuum anneals, the final autoclave treatment, and the final inspection.

The fabrication routes, Figure 1, were devised on the basis that: a) the texture of pressure tubes is mainly a function of extrusion ratio, and a minor function of the billet microstructure and the post extrusion fabrication process/4/, and b) dislocation density and grain shape are dependent on the extrusion conditions, and the amount of post extrusion cold work and heat treatment/5/.

The use of a lower extrusion ratio than for current tubes was designed to produce a less intense circumferential texture, but the reduction was limited by restrictions on billet geometry and the amount of cold reduction following extrusion. Also press capacity limited the reduction in extrusion temperature to 40 K less than normal thus limiting the ability to take advantage of favourable strength and texture changes by extruding at lower temperatures.

Route 1 was designed to produce strengthening from additional cold work but with a lower dislocation density achieved by a higher stress relief treatment than in current tubes.

Route 2 was designed to produce a less elongated grain shape than Route 1 by an anneal halfway through the cold working.

Route 3 was designed to produce a modified grain shape by annealing after extrusion, with the grain shape and dislocation density subsequently modified by cold working and further heat treatment.

Each step of the fabrication sequence proceeded without significant difficulty. Double consumable arc melting of a 5860 kg ingot produced 34 billets, of which 28 were used in this program. Examination of slices from the forged billets by eddy current techniques, showed that there was no significant segregation of oxygen or niobium. Instrumented hollow billets were used to establish the conditions for the beta treatment of the billets. Macro and microstructural examination of billet slices showed a uniformly small prior beta grain structure and except for a thin martensitic surface layer, a uniform alpha grain structure.

All extrusion conditions including billet heating cycles were closely controlled and except for one shell, a product typical of standard production quality was produced. Cold drawing of the extrusions required no special techniques, although the 40% reduction on Routes 1 and 3 were significantly higher than the typical 27% in current production. Intermediate heat treatments were performed in a vertical single retort vacuum furnace with the heat up and cool down cycle closely maintained and the temperature over the tube length controlled within ± 15K. Each heat treatment condition was determined from prior tests on current Zr-2.5% Nb tubing and confirmed by the tests on the relevant tube (Appendices D and E).

3.0 RESULTS

The tubes were evaluated from two aspects:- a comparison with specification requirements and a more intensive investigation into their properties including in-reactor testing and condition after assembly into end fittings.

3.1 Chemical Analysis

The ingot analysis is compared to current Zr-2.5 Nb in Table 1. The specifications restricted the niobium concentration to the high end of the currently specified range and required the oxygen to be higher. These requirements were met and in other respects the analysis was typical of current material. Check analyses on finished tubes showed oxygen concentrations averaging 1330 ppm.

3.2 Tensile Properties

For design purposes pressure tubes must satisfy a minimum 573 K UTS value of 480 MPa, as measured on longitudinal specimens machined from the front end of each tube. However, for this program specimens were machined from the front and back ends* of each pressure tube and the specimens tested using the specified testing procedures, Table 2. All tubes satisfied the design requirements and were significantly stronger than current tubes. The Route 3 tubes were much stronger than required and they were given an additional stress relief at 748 K for six hours prior to autoclaving.

The influence of neutron (E>1 MeV) irradiation on the room temperature and 573 K tensile properties of a tube from each route has been investigated. Irradiation at 573 K to a fluence of

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*the front of a tube is the end which is extruded and cold drawn first.

1.6×10^{24} n/m^2 increased their strength and decreased their ductility similar to that observed in current tubes, Table 3.

3.3 Corrosion

For design purposes, pressure tubes are corrosion tested in accordance with the ASTM G-2 tests. All the specimens had weight gains less than the maximum allowed for CANDU tubes, Table 4. Further corrosion tests were performed in an experimental loop in the NRU reactor at AECL, Chalk River. The specimens were exposed to a fast (E>1 MeV) neutron flux and primary heat transport water conditions. Table 5 records the weight gains where it is observed that Route 2 and 3 materials have lower weight gains than current tube material, (the results of Route 1 at the higher fluences are not available as specimens are still in the reactor).

Corrosion tests were also performed to determine the susceptibility of the different tubes to crevice corrosion. From each tube two corrosion coupons (17 x 10 x 4 mm) were machined. The coupons were placed in an autoclave with the following test conditions:

Concentration of LiOH	1.0 M
Temperature	340°C
Pressure	14.8 MPa
Duration	4 days

Table 6 records the weight gains and hydrogen uptake. The results indicate that Route 2 and current tubes have similar weight gains and hydrogen uptake. However, Route 1 and 3 tubes have lower weight gains but higher hydrogen uptake than current tubes. From the point of view of crevice corrosion Route 1 and 3 tubes should have an advantage over Route 2 or current tubes.

3.4 Hydride Phenomena

3.4.1 Hydrogen Diffusion & Terminal Solid Solubility (TSS)

Four coupons 45 mm long x 15 mm wide were machined from the back ends of each tube. A hydride layer was electrolytically formed on one end of each coupon. Two coupons from each tube were annealed at 523 ± 2 K for 430 h; the other two coupons from each tube were annealed at 573 ± 2 K for 140 h. Optical metallography and hydrogen analyses were both used to determine the hydrogen profiles along the length of each coupon. These results were analysed by finding either a two or three-parameter fit to the standard solution of the diffusion equation. The terminal solid solubility was obtained as one of the parameters in the solution to the diffusion equation. The results are presented in Table 7. The differences in the diffusion coefficients in the different tube materials are small although they appear to be significant. Route 2 material has diffusion coefficients at 523 and

573 K which are not significantly different from those of a current tube. However, Route 1 and 3 tubes have similar diffusion coefficients which are significantly lower than those for a current tube. The differences in the hydrogen diffusion coefficient are unlikely to have a significant effect on hydride distribution in TG3 tubes as compared to current tubes. The time to reach TSS in all tubes is expected to be similar and there is no significant difference in the TSS of the tubes (Table 7).

3.4.2 Stress to Reorient Hydrides

Two experimental techniques were used:

1) Ring Specimens: Transverse rings were cut from the tubes, hydrided to 100 ppm hydrogen and stressed on D grips, Figure 2. The wall thickness of the rings was reduced in five places to give different stress zones. The rings were positioned with these zones in the middle of the 'D' so that when the load was applied the stress in the zones was circumferential. After the load was applied the rings were heated to 573 K for two hours and the furnace-cooled to room temperature. The transverse faces of the stress zones were examined metallographically and the orientation of the hydrides measured.

In the as-fabricated condition the hydrides in all tubes are oriented within 10° of the circumferential direction. In all the tubes a circumferential stress of 280 MPa oriented the hydrides 30-60° from the circumferential direction towards the radial direction. Increasing the stress to 420 MPa oriented the hydrides close to the radial direction, Table 8. There was no significant difference between the tubes.

2) Tapered Tensile Specimens: Two tapered tensile specimens were made from straightened material from each of the front and back ends of one tube from each route. The straightened material had been stress relieved 24 hours at 673 K and then electrolytically hydrided. Specimens were tested in a constant load creep machine at a load level which produced a stress distribution from 100 to 500 MPa along the specimen length. Specimens were heated and held under load at 573 K for 48 hours before furnace cooling at a rate of 1.5 K/minute. Following cooling, the specimens were examined metallographically on a radial-transverse plane. Analysis of the hydride orientation distribution as a function of stress, produced values of the threshold stress for formation of completely radial hydrides. Two different types of analysis were made: the results from both analyses are shown in Table 9. The results indicate that the back ends have a higher threshold stress than the front ends in all routes and that Route 2 material appears to have a slightly higher threshold than material from the other two routes. Tests of material from the back end of a current pressure tube produced results which were comparable with those obtained at the front ends of the TG3 materials.

There appeared to be a correlation between increasing the tube strength and the stress to produce radial hydride which is discussed more fully in Appendix E.

The stress at which some radial hydrides first appear in these materials was also estimated. Material from Routes 1, 2 and the current tube exhibited some radial hydride at stresses as low as 175 MPa whereas Route 3 material had radial hydride at 150 MPa.

3.4.3 Delayed Hydride Cracking

Two experimental techniques were used:

1) Cantilever Beam Specimens: The susceptibility of the tubes to crack initiation by delayed hydride cracking/6/ was assessed using cantilever beam specimens, Figure 3. Specimens were hydrided to 50 ppm hydrogen. One set of specimens was loaded to an outer fibre stress of 620 MPa, heated to 575 K and cooled to a test temperature of 525 K. A second set of specimens was loaded to an outer fibre stress of 550 MPa, heated to 575 K and cooled to a test temperature of 425 K. Once a week all specimens were cooled to room temperature and reheated to the test temperature. The results summarized in Table 10 show that under very severe test conditions Routes 1 and 2 are more susceptible to delayed hydride cracking than Route 3, and current tubes. Under less severe conditions, sensitivity of TG3 tubes to cracking is similar to current tubes.

2) Compact Tension Specimens: Small compact tension samples were prepared from material from the back end of one tube from each route. The material was straightened, stress relieved 24 hours at 673 K, electrolytically hydrided, diffusion annealed at 523 K to produce about 30 ppm of hydride, machined, fatigue-precracked and stress relieved again at 673 K for 24 hours. The test procedure consisted of loading the specimens to produce a nominal stress intensity factor at the fatigue crack tip of about 6 MPa $\sqrt{m}$, with crack growth in the axial direction. The specimens were then heated to 573 K and cooled at a rate of 0.33 K/minute to the test temperature (483 K or 423 K). If no cracking initiated within a 24-hour period, the load was increased to change the stress intensity factor at the crack tip by about 0.75 MPa$\sqrt{m}$. Another 24-hour period was allowed before any further load increments were made. When cracking initiated, the load was kept constant and after a suitable cracking period, the test was terminated. The crack velocity was determined from the average crack extension and the cracking time. Table 11 shows the range of results obtained for each material as well as results obtained previously on current Zr-2.5 Nb. For both test temperatures the values of K_{IH} and crack velocities for the TG3 materials fall within the range of values quoted for current Zr-2.5 Nb. Tests at 483 K were performed on all four materials subjected to the same preparation and test techniques. These results indicate

that TG3 materials have similar K_{IH} values and crack velocities to the current Zr-2.5 Nb.

3.5 Flaw Tolerance

Sections of the tubes 460 mm long had about 100 ppm hydrogen added. In each section a longitudinal slit was spark machined through the wall and extended by fatigue cycling. Aluminum liner tubes were inserted inside the sections and the assemblies internally pressurized to determine the pressure required for unstable extension of the flaws/7/. The results are summarized in Figure 4 and compared with the lower bound line for current tubes/8/.

At room temperature the stresses required to propagate the flaws were higher than the lower bound line for standard tubes and at 570 K their toughness was similar to current tubes.

3.6 Wear Properties

Wear tests were performed in an autoclave at about 538 K. A standard fuel bundle bearing pad was held against the pressure tube specimens with a static normal force of about 1 N. The bearing pad was then vibrated against the fixed pressure tube specimens. Two sets of test conditions were used: a) vibration at 18 Hz with peak to peak amplitude of 0.2 mm and b) vibration at 21 Hz with peak to peak amplitude of 0.4 mm. Both the pressure tube specimen and the bearing pad were weighed before and after each test. The results of these tests are presented in Table 12.

3.7 Rolled Joint Properties

Rolled joints made with TG3 pressure tubes were tested to determine their characteristics relative to current rolled joints. Modified zero clearance rolled joint procedures were used to make the test assemblies. Helium leak tests were performed on all the joints, pull out tests were performed on double-ended assemblies and residual stresses were measured on single-ended assemblies. The results are summarized in Table 13 and details are presented in a summary report contained in Appendix E. A detailed report on the rolled joint evaluation studied will be issued separately by AECL-CANDU Operations/9/.

3.8 Microstructure

Evaluation of the metallurgical structure was concerned with: i) the uniformity of the structure between tubes fabricated by the same route; ii) the variation between the front and back of each tube, and iii) the differences between TG3 tubes and current tubes.

3.8.1 Grain Shape and Phase Distribution

Current Zr-2.5 wt% Nb pressure tubes have a microstructure that consists of elongated α_{Zr} grains with a continuous grain boundary network of retained β_{Zr}. Figures 5(a) - 5(d) are typical scanning electron micrographs of longitudinal sections from the front ends of each type of pressure tube and show the difference in the distribution and morphology of the β-phase between the different tubes. In all tubes the β-phase is elongated in the axial direction and outlines the alpha grains. Route 2 and 3 tubes have a coarser β-structure than Route 1 or current tubes. The α-grain shape is more clearly observed in the transmission electron micrographs (Figures 6(a) - (d)). Route 1 material has a similar α-grain shape to current material, ie highly elongated (Table 14), with a continuous grain boundary network of β-phase. In Route 2 and 3 tubes the α-grains are less elongated and the β-phase is not continuous along the grain boundaries.

Qualitative observations from the scanning electron micrographs indicate that the tubes manufactured by a given route have similar microstructures. In each route the alpha grain thickness is smaller at the back ends than at the front ends of the tubes.

3.8.2 Dislocation Density

X-ray line broadening techniques were used to measure the dislocation density of material from the front and back end of each tube (Table 15). Route 1 and 3 tubes have a lower dislocation density than Route 2 tubes which is similar to current tubes. For a given route the tube-to-tube variation is small.

3.8.3 Texture

The texture was measured by X-ray diffraction techniques which determines the resolved fraction of basal poles in the principal directions (f_d) of the tube/10/. Typically cw Zr-2.5 Nb tubes have a crystallographic texture with the basal poles concentrated in the circumferential direction. Table 16 records the average f_d values for the front and back of the tubes manufactured by the modified routes and typical values for current tubes. The tube-to-tube variation in texture is small within each route. There is also a small difference in texture between the front and back ends of each type of tube.

For Route 2 and 3 tubes the texture was influenced by the annealing temperature (Table 17). As the annealing temperature is increased the resolved fraction of basal poles in the circumferential direction is increased.

3.9 In-Reactor Dimensional Changes

3.9.1 Irradiation Creep

In-reactor creep rates of each type of pressure tube were determined from bent beam stress relaxation tests/11/. The specimens

were irradiated to a neutron (E>1 MeV) fluence of 5.3×10^{24} n/m^2 at about 530 K with an initial fibre stress of 190 MPa. The relaxation rate constants are presented in Table 18. There is no significant difference in relaxation rate constants, and hence, in the irradiation creep rates of any of these tubes.

3.9.2 Irradiation Growth

This program was performed in the DIDO reactor at AERE Harwell at a neutron (E>1 MeV) flux of about 6.5×10^{17} n/m^2s and at 553 K. Longitudinal and transverse specimens were machined from the middle of one tube from each route and a current tube. The specimens were in the cold worked condition so that after machining they could be stress relieved under the relevant conditions for the specific routes. All the specimens were stress relieved while clamped flat. The specimen dimensions are 60 mm x 9.9 mm x 1.5 mm. Length measurements are made to an accuracy of $\pm$ 0.5 µm.

A single tier of the irradiation rig enables twenty specimens to be irradiated at a given time. Batches of specimens were rotated in the rig and after each irradiation exposure their length was measured. Figure 7 presents the growth characteristics of longitudinal and transverse specimens from the different pressure tubes. For all tubes the transverse specimens exhibit a continuous decrease in length with neutron (E>1 MeV) fluence and Route 2 tubes have the lowest growth rate. The longitudinal specimens all exhibit initial length increases. However, the subsequent longitudinal growth behaviour differs for each type of material. Route 1 material exhibits a decrease in length; current and Route 2 materials remain relatively constant before exhibiting an increase in growth rate; Route 3 material shows a constant increase in length with neutron (E>1 MeV) fluence.

4.0 DISCUSSION

The three fabrication routes were designed to make Zr-2.5 Nb tubes that had a different crystallographic texture, grain shape and dislocation density to current tubes. Qualitatively, this objective was achieved and the differences between the tubes and current tubes are summarized in Table 19. The program also showed that there is considerable flexibility in manufacturing tubes to meet current specifications and quality requirements.

Each of the routes produced tubes with tensile strengths and ductilities above those of current tubes. Consequently, the modified tubes satisfy the design tensile requirements for reactor installation.

Strength of tubes with post extrusion heat treatments had been a concern, hence the specified niobium and oxygen concentrations. However, the concentrations of those elements are only slightly higher than in current material and unlikely to be the source of

the strengthening observed. The strength of Zr-2.5 Nb has been seen/4/ to be linearly dependent on the inverse root of the grain size, yet in Routes 2 and 3 the grain size was much thicker(larger) than in current tubes. However, there is evidence/12/ that the 475-500°C S/R treatments promote subgrain formation and transformation and reorientation of the β-phase. These microstructural changes are considered to be the reason for the higher than expected strength of the modified tubes. The higher strength at the back ends of the tubes is attributed to the smaller grain size.

A higher tensile strength is of no advantage for in-reactor deformation. Hence Route 2 tubes could also have been stress relieved before autoclaving to reduce their tensile strength and dislocation density and Routes 1 and 3 tubes could have been stress relieved at higher temperatures. The higher strength of the modified tubes did not continue to be an advantage in the irradiated condition.

All the routes had lower weight gains in the ASTM - G-2 corrosion test than is specified for CANDU pressure tubes. The evaluation corrosion tests, performed in and out-reactor, showed that the TG3 tubes had lower weight gains than current Zr-2.5 Nb. The results from the longer term irradiation tests will be included in Appendix E when they are available. Under crevice corrosion conditions the weight gains of Route 1 and 3 tubes was lower than that of Route 2 and current tubes. For the severe crevice corrosion test conditions the hydrogen pick-up tends to be higher in the tubes with the lower weight gains.

From the results discussed above, the modified pressure tubes satisfy all the design requirements for installation in power reactors.

The pressure tubes contain 10-20 hydrogen when they are installed in a reactor and are predicted to pick up about 80 ppm deuterium during thirty years' service. Hence after thirty years' service the equivalent hydrogen concentration of the tubes will be about 50 ppm. At room temperature the solubility of hydrogen in zirconium is essentially zero/13/ and the tubes contain zirconium hydride platelets about 0.01 mm diameter/14/. When the tubes contain >50 ppm hydrogen, hydride platelets will be present at reactor operating temperatures (523-585 K). In as-manufactured pressure tubes the hydrides are oriented on the circumferential-axial plane and their effect on mechanical properties is small. However, a circumferential stress and a thermal cycle to 523 K can reorient the platelets onto the radial-axial plane and then they lower the ductility of the tubes. The maximum circumferential stress in a pressure tube in a CANDU reactor is 200 MPa/8/, and the transverse ring tests show that none of the tubes would have any hydrides oriented on the radial-axial plane. This result is confirmed by the tapered tensile tests. Based on the residual stress balancing analysis these tests show that hydride reorientation occurs at stresses greater than 200 MPa for all tubes. The tapered tensile test data indicates that the stress to re-orient hydrides in TG3 tubes is greater than in current tubes.

Although not a safety requirement, additional confidence in the operation of CANDU pressure tubes comes from the leak-before-break criterion. For leak-before-break to occur, it is necessary to have a sufficiently long critical crack length so that a leak will occur before any cracks become unstable. The slit burst tests show that the critical crack length of the TG3 tubes was no worse than that for current tubes at 575 K and was probably better at room temperature. Hence the tubes should have a similar flaw tolerance to current tubes. Since the transverse stress from the coolant pressure is twice the longitudinal stress the most severe flaw is a longitudinal crack. The most probable mechanism for forming a crack is delayed hydride cracking. Cracks can initiate by this mechanism at small flaws at stresses below the ultimate tensile strength and grow until they penetrate the wall of the tube/15/. The cantilever beam tests showed that there was little difference in susceptibility to hydride cracking between these tubes and current tubes. We assume that the tubes would also have a similar resistance to delayed hydride cracking at the much lower stresses in a power reactor.

The cantilever beam results are supported by the data generated from compact tension specimens (Table 11). TG3 tubes have comparable K_{IH} values to current tubes which have values reported in the range 6-9 MPa$\sqrt{m}$. The crack velocity data is temperature dependent with lower velocities at lower test temperatures. For current tubes there is a large range of velocities at a given test temperature. The TG3 crack velocity data falls within the range of values reported for current tubes. For a test temperature of 583 K the crack velocity measured in a current tube is higher than that of TG3 tubes. These tests were performed on specimens subjected to the same preparation and test techniques, which is not necessarily the case for tests on current tubes.

The results of the wear tests (Table 12) illustrate that each TG3 tube has similar wear characteristics for both test conditions. Comparison with a current tube shows that the TG3 tubes have better wear properties.

The leak rates of rolled joints made at site but out-of-reactor must satisfy the specified maximum of 2.0×10^{-8} cc/s He. All the TG3 rolled joints satisfy this criterion (Table 13). For rolled joints made in-reactor the specified maximum leak rate is 3.0×10^{-6} cc/s He. This condition is satisfied with TG3 rolled joints even after thermal cycling where the range of leak rates is 2×10^{-8} - 1.24×10^{-6} cc/s He. The results of the pull-out strength and residual stress measurements on TG3 rolled joints are consistent with results on current rolled joints.

The main purpose of the modified routes was to change the metallurgical structure of the pressure tubes so as to reduce the in-service elongation rates. An equiaxed grain shape is expected to reduce the irradiation growth rate because of a reduction in the anisotropy of the distribution of the grain boundaries which act as neutral sinks/16,17/. The production of a pressure tube with

equiaxed grains and adequate strength is not practical. Consequently, the approach taken was to reduce the aspect ratio of the grains by a re-crystallization anneal in Routes 2 and 3. The scanning electron micrographs (Figures 5(a)-(d)) show that the β distribution and size were also changed by the anneals. In the Route 1 tubes the "break up" of the β was indicative of the transformation of the metastable β_{Zr} to the β_{Nb}/18/.

The difference in texture between the modified and current tubes is in the correct direction to reduce elongation rates. However, the magnitude of the change is much less than was predicted from the studies on Zircaloy pressure tubes/2/. The ratio f_R/f_T is used in predicting elongation rates and the "ideal" would be a completely random texture, ie $f_d = 0.33$. The difference in texture between the routes indicates that the post-extrusion processes influence texture. Route 1 tubes have the best texture since an adverse texture rotation was observed on annealing in Routes 2 and 3 (Table 14) which recovered on further drawing. In all the tubes the higher value of f_R/f_T at the back ends than at the front ends probably relates to the decrease in temperature during the extrusion of a tube. To increase the ratio to more than 0.67 the extrusion ratio would have to be further reduced by either using a smaller diameter extrusion billet or by extruding a thicker walled tube that would require more than 40% reduction to the final size. The irradiation creep contribution to in-service deformation is primarily controlled by the crystallographic texture and only has a weak dependence on grain shape and dislocation density. From the in-reactor stress relaxation results all the tubes are predicted to have similar in-reactor creep rates which probably reflects the small difference in texture between tubes.

The lower dislocation densities in Routes 1 and 3 reflect the high temperature stress reliefs. A reduction in dislocation density reduces the rate of irradiation growth in Zircaloy and Zr-2.5 wt% Nb/19,20/. For irradiation growth the texture determines the direction of growth but the grain shape and dislocation density are expected to control the growth rate. The growth characteristics of the Route 1 tube after the initial strain increase is unusual as the decrease in length is not consistent with the texture. However, this effect is considered to be transient/21/. For the Route 1 tube the growth results from the longitudinal and transverse specimens indicate that an expansion in the wall thickness must be occurring to maintain constant volume.

5.0 CONCLUSIONS

1. The Task Group 3 development program has demonstrated that different fabrication routes can be used to produce pressure tubes which satisfy the quality assurance and design requirements for installation in CANDU nuclear reactors.

2. The TG3 pressure tubes exhibit similar hydride cracking characteristics and flaw tolerance values to current Zr-2.5 wt% Nb pressure tubes.

3. The only significant difference between TG3 tubes and current tubes is in their irradiation growth behaviour. All three TG3 routes produce tubes with lower irradiation growth rates than current tubes. This result indicates that the elongation rates of the TG3 tubes will be lower than current tubes and Route 1 tubes should have the lowest elongation rates of all the tubes.

4. The tensile strength of the TG3 pressure tubes is not a simple function of dislocation density or grain size, but is related to the sub-grain size and the distribution of the 'β' phase.

5. If pressure tubes with elongation rates lower than current tubes are required then a TG3 production route should be used.

6.0 RECOMMENDATION

Irradiation tests on specimens from Zr-2.5 Nb tubes indicate that Route 1 tubes should have the lowest in-service elongation of all the tubes tested. The TG3 development program has shown that Route 1 produces tubes which satisfy all the quality assurance requirements for reactor installation. The Route 1 production process should be considered as an alternative to the current process for the production of future pressure tubes.

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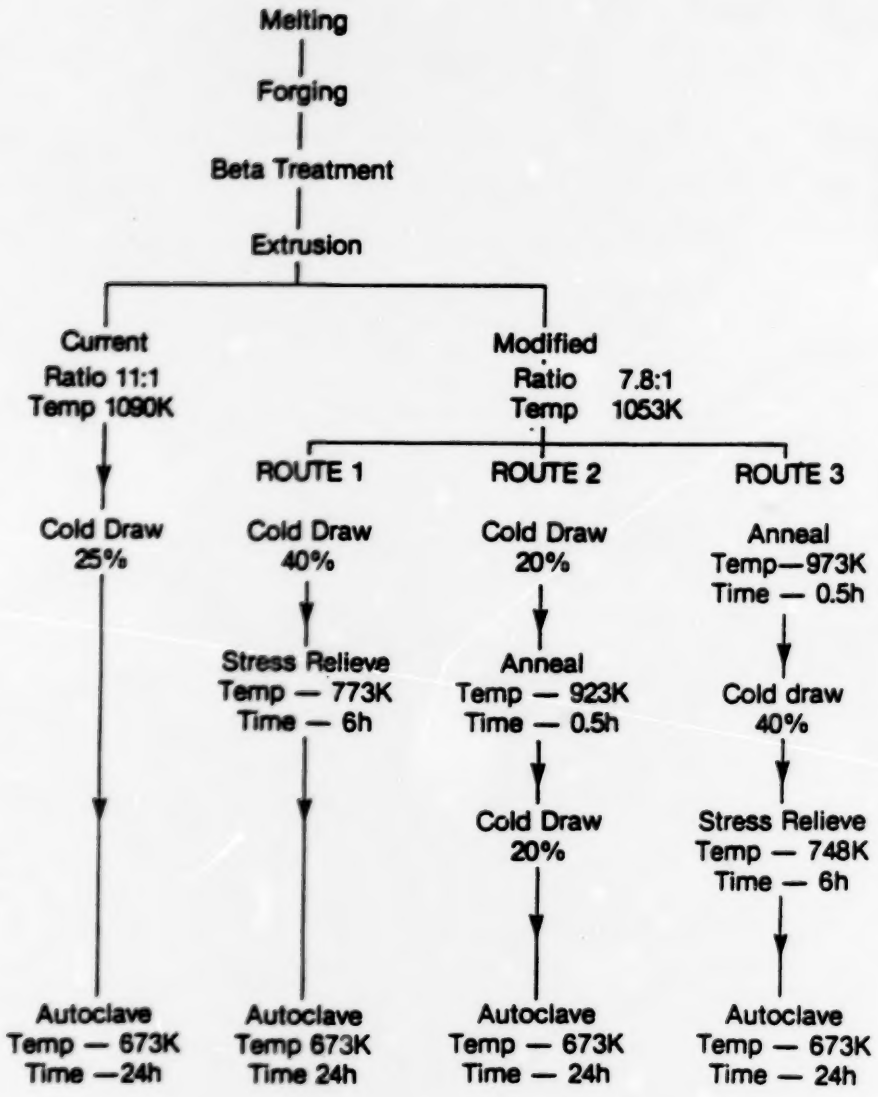
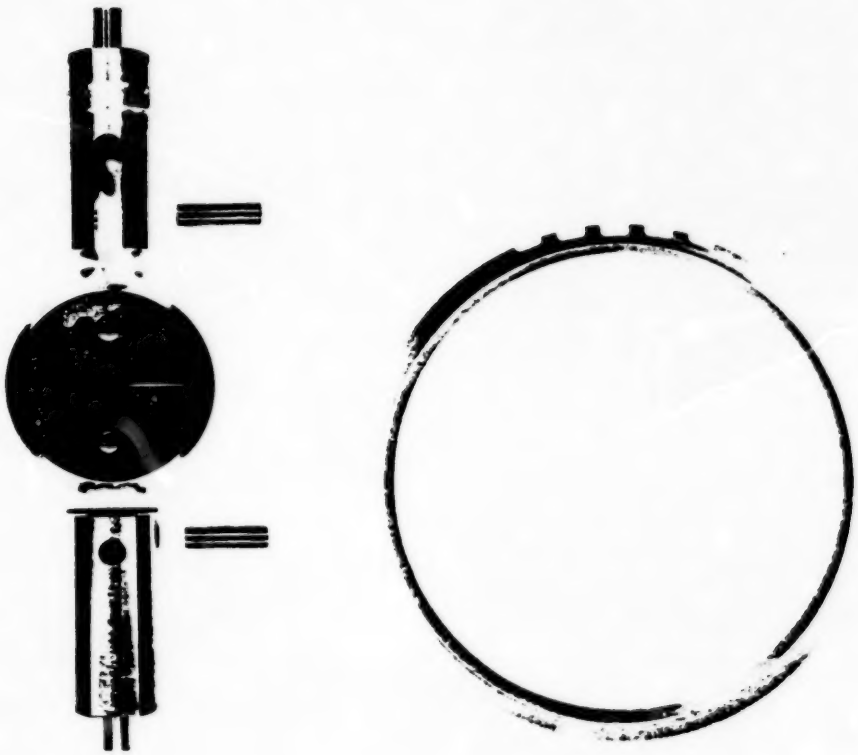


FIGURE 1

PRODUCTION ROUTES FOR THE FABRICATION OF COLD WORKED Zr-2.5 wt% Nb TUBES



a b

(a) D-grip Jig for Stressing Rings
(b) Detail of Slotted Ring Specimen

FIG 2 - HYDRIDE ORIENTATION TESTS

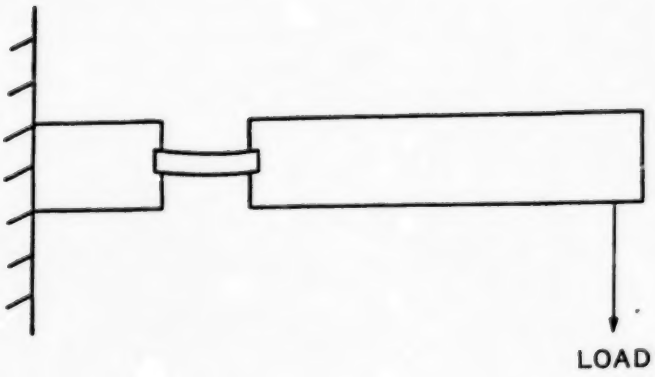


FIG 3 - SCHEMATIC OF THE CANTILEVER BEAM SPECIMEN TEST ON SECTIONS OF TUBES

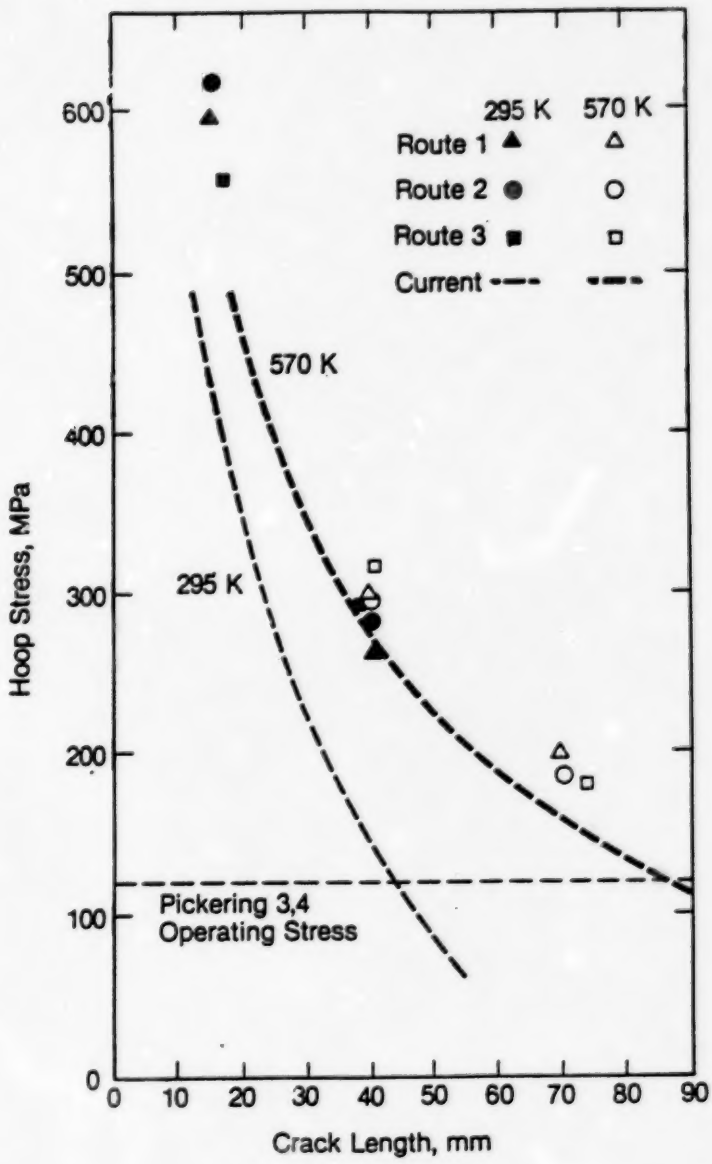
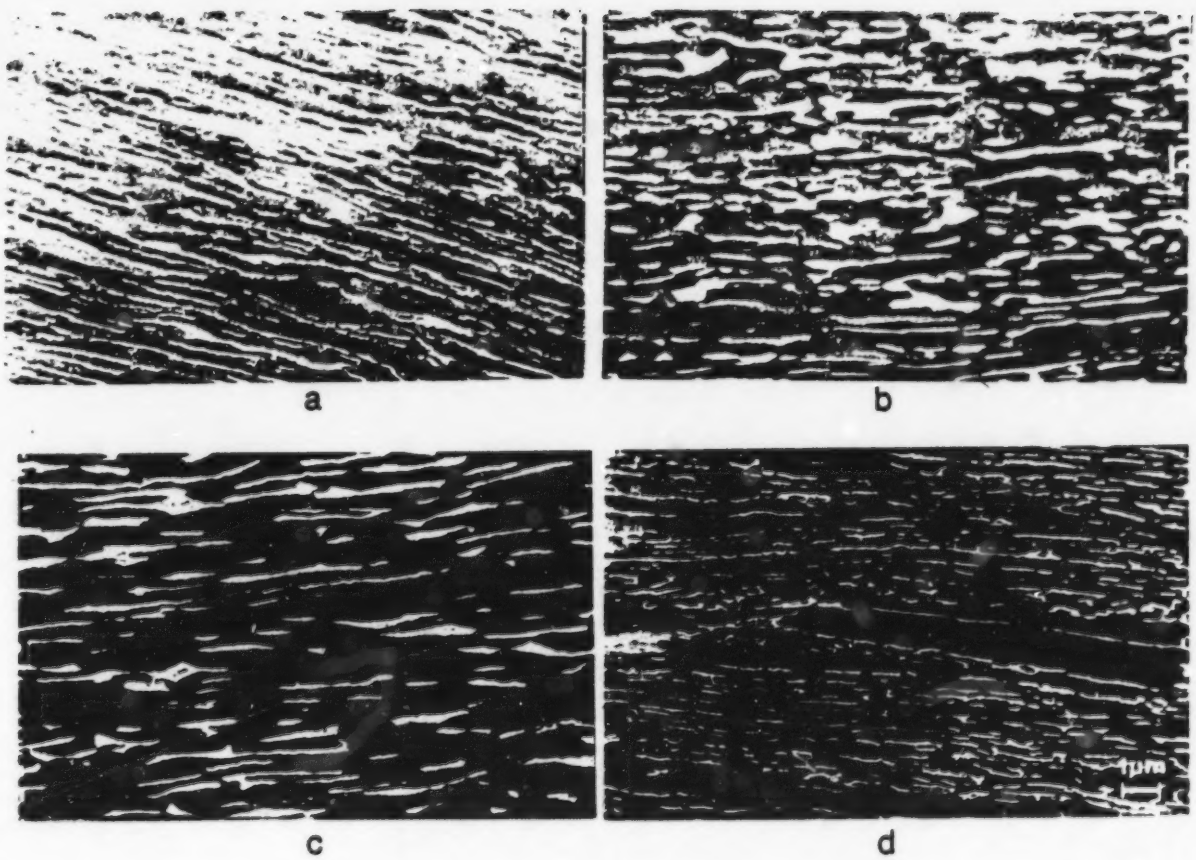


FIGURE 4

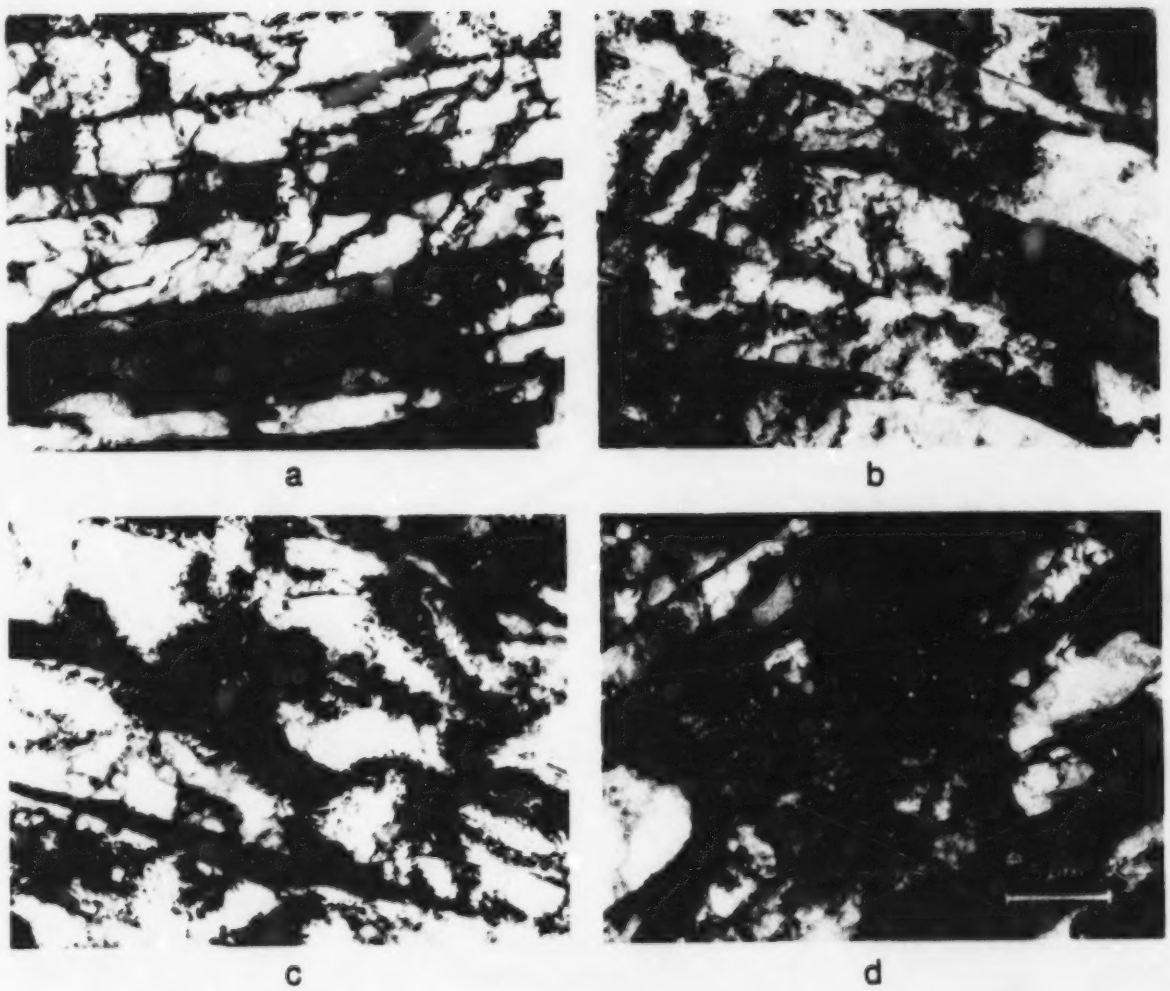
HOOP STRESS REQUIRED TO PROPAGATE A THROUGH-WALL CRACK IN Zr-2.5 wt% Nb TUBES FABRICATED BY THREE NEW ROUTES



(a) Route 1 tube
(b) Route 2 tube
(c) Route 3 tube
(d) Current tube

FIG. 5 *Scanning electron micrographs of tubes on axial-radial sections*

01-00822-5964



(a) Route 1 tube
(b) Route 2 tube
(c) Route 3 tube
(d) Current tube

FIG. 6 *Transmission electron micrographs through axial-radial plane of tubes*

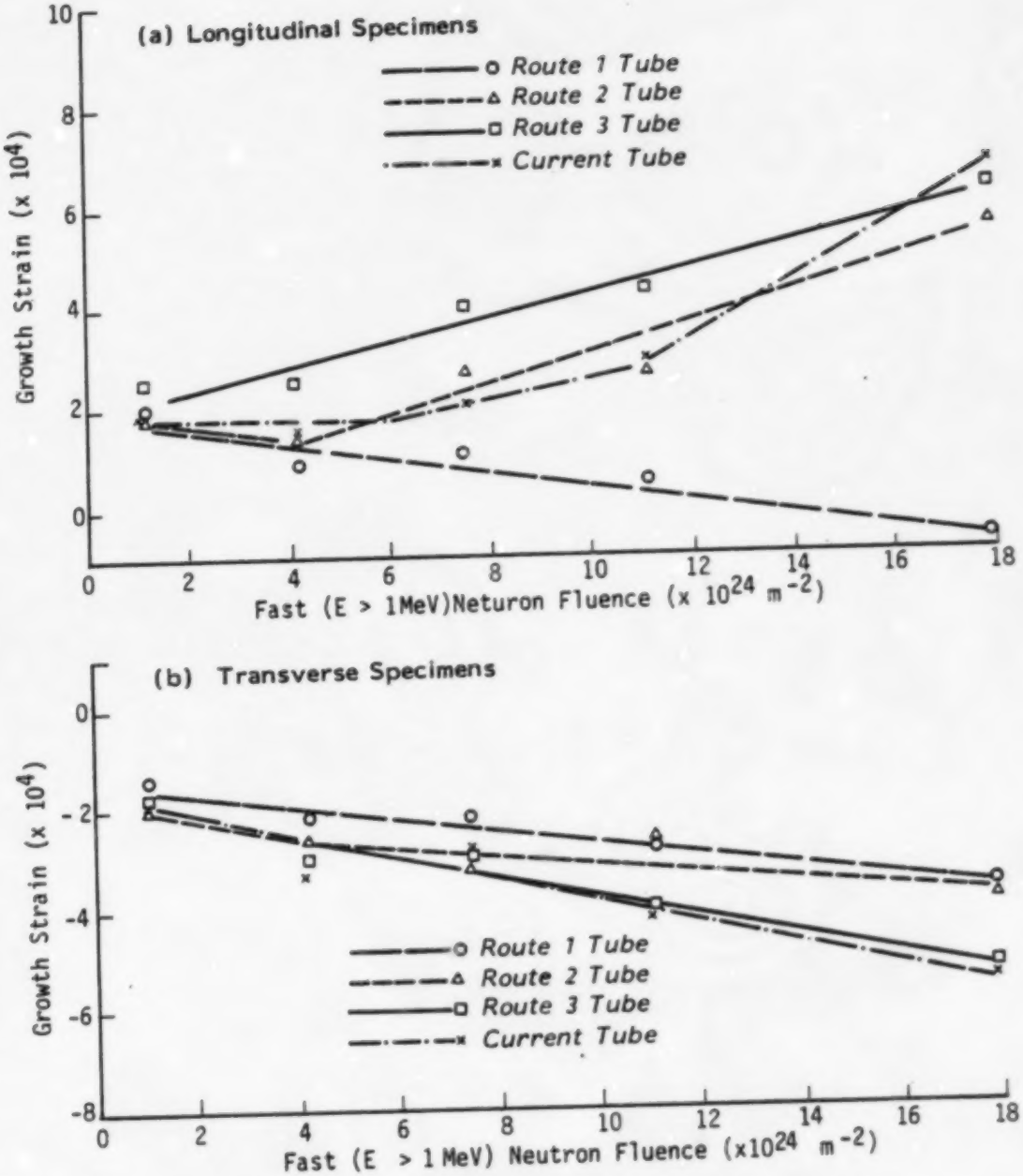


FIGURE 7

THE IRRADIATION GROWTH STRAIN AS A FUNCTION OF FAST (E>1 MeV) NEUTRON FLUENCE FOR TG3 AND CURRENT PRESSURE TUBE MATERIALS

TABLE 1

CHEMISTRY REQUIREMENTS AND ANALYSES

Current c.w. Zr-2.5 wt% Nb			Modified c.w. Zr-2.5wt% Nb		
Element	Requirement (% or ppm)	Typical Range	Requirement (% or ppm)	Analysis Ingot	Analysis Tube
Nb	2.4-2.8	2.5-2.7	2.7-2.8	2.64	2.71
O	900-1300	1100-1200	1200-1600	1490	1330
N	65 max.	<20-55	65 max.	43	30
H	20 max.	< 5-16	20 max.	< 5	9

TABLE 2

SUMMARY OF TENSILE PROPERTIES OF MODIFIED AND CURRENT c.w Zr-2.5 wt% Nb TUBES AT 573K

Route		U.T.S (MPa)		0.2% YS (MPa)		Elongation % in 4.5 $\sqrt{A}$	
		Front	Back	Front	Back	Front	Back
1	Mean	543	569	371	399	18	18
	Range	534-559	564-574	354-379	343-405	16-24	15-21
2	Mean	541	560	401	412	17	18
	Range	530-548	569-565	390-408	408-418	14-20	16-19
3	Mean	538	542	383	382	17	19
	Range	532-544	536-547	363-391	379-386	14-20	18-20
3 Without 728K S/R	Mean	576	586	481	490	16	17
	Range	569-586	561-602	463-492	478-508	14-18	16-18
Current Specification For Zr-2.5 wt% Nb Tubes							
	Mean	516	--	394	--	16	--
	Range	482-558	--	351-338	--	12-20	--

TABLE 3

EFFECT OF NEUTRON IRRADIATION ON THE LONGITUDINAL TENSILE PROPERTIES OF THE c.w. Zr-2.5 wt% Nb TUBES

Fabrication Route	Test Temperature K	Neutron Fluence 10^{24} n·m^{-2}	0.2% YS (MPa)	UTS (MPa)	Elongation %
1	293	0	574	788	15
	293	2.0	808	977	10
	573	0	360	537	17
	573	1.6	641	735	13
2	293	0	603	794	14
	293	2.0	897	1003	8
	573	0	401	543	19
	573	1.6	655	758	10
3	293	0	581	792	16
	293	2.0	785	983	10
	573	0	388	541	16
	673	1.6	657	742	10
Current Tubes	293	0	545	747	13
	293	-	---	---	--
	573	0	433	535	14
	573	1.6	702	756	7

TABLE 4

CORROSION WEIGHT GAINS AFTER THE ASTM G2 CORROSION TEST

Fabrication Route	Weight Gain (mg/dm^2)
1	18 - 21
2	26 - 30
3	23 - 26
Current Tubes	20 - 37
Specifications	35

TABLE 5

CORROSION WEIGHT GAINS FOR SPECIMENS EXPOSED TO A FAST (E 1 MeV) NEUTRON FLUX

Fabrication Route	Weight Gain (mg/dm^2)
1	
2	13 - 17
3	17 - 20
Current	32 - 33

TABLE 6

WEIGHT GAIN AND HYDROGEN UPTAKE FOR SPECIMENS EXPOSED TO ACCELERATED CREVICE CORROSION CONDITIONS

Fabrication Route	Weight Gain (mg/dm^2)	Hydrogen Uptake ($\mu g/g$)
1	898 - 904	33
2	1104 - 1163	6
3	804 - 809	26
Current	1190 - 1196	10

TABLE 7

DIFFUSION COEFFICIENTS & TERMINAL SOLID SOLUBILITIES (TSS) OF THE TUBES

Fabrication Route	Temperature (°C)	Diffusion Coefficient x 10^{n} m^2s-1	TSS µg/g
1	250	3.94	29.7
	300	7.21	54.6
2	250	5.02	31.7
	300	12.6	54.8
3	250	4.13	26.1
	300	8.08	54.2
Current	250	5.02	34.5
	300	13.2	52.4

TABLE 8

THE EFFECT OF CIRCUMFERENTIAL STRESS AND A THERMAL CYCLE TO 575 K ON THE ORIENTATION OF HYDRIDES IN THE TUBES

	Typical Angle Between Reoriented Hydrides And The Circumferential Direction			
Stress MPa	*Route 1*	*Route 2*	*Route 3*	*Current*
0	*0-10*	*0-10*	*0-10*	*0-10*
280	*30-60*	*30-70*	*30-70*	*50-60*
420	*50-90*	*50-90*	*50-90*	*70-90*

TABLE 9

STRESS REQUIRED FOR 100% RADIAL HYDRIDE

Material	Stress* By 50% of Wall Criterion	Stress* - By Residual Stress Balancing
Route 1 Back	218	224
Front	195	208
Route 2 Back	229	241
Front	217	216
Route 3 Back	218	211
Front	198	203
Bruce Tube-Back	193	211

* Techniques Used To Obtain These Values Described in Appendix

TABLE 10

SUMMARY OF THE CANTILEVER BEAM
DELAYED HYDRIDE CRACKING TEST ON THE TUBES

Tube	Stress MPa	Test-Temperature K	Failure Times h	Specimens Still on Test h
Route 1			150-10,830(8)	All Failed
Route 2	620	525K	320-1,850(8)	All Failed
Route 3			485-8,065(6)	19,010(2)
Current			1,325-8,700(5)	15,145(3)
Route 1			-	15,815(3)
Route 2	550	425(k)	-	15,815(3)
Route 3			-	15,815(3)
Current			-	15,815(3)

*( ) Number of Specimens

TABLE 11

K_{IH} AND VELOCITIES FOR DELAYED HYDRIDE CRACKING

Route	Test Temp (°C)	K_{IH} MPa $\sqrt{m}$	Velocity m/s
1	210	6.5-7.1	$1.4\text{-}1.5 \times 10^{-8}$
2	210	6.5-7.3	$1.5\text{-}1.8 \times 10^{-8}$
3	210	6.4-6.5	1.8×10^{-8}
Current	210	7.1-7.4	$2.9\text{-}3.0 \times 10^{-8}$
Results From Literature (Current Tube Material)	210		1.2×10^{-8} $+2\sigma$ 3.16×10^{-8} -2σ 4.7×10^{-9}
1	150	6.7-7.5	$2.3\text{-}2.6 \times 10^{-9}$
2	150	6.8-6.9	$2.8\text{-}3.1 \times 10^{-9}$
3	150	6.0-6.1	$1.7\text{-}1.9 \times 10^{-9}$
Results From Literature (Current Tube Material)	150		1.99×10^{-9} $+2\sigma$ 5×10^{-9} -2σ 7.7×10^{-10}

TABLE 12

RESULTS OF WEAR TESTS ON PRESSURE TUBE/
FUEL BUNDLE BEARING PAD ASSEMBLIES

Material	*Weight Loss Ratio*	
	Pressure Tube/Bearing Pad	
	18 Hz 0.2 mm	*21 Hz 0.4 mm*
Route 1	*2.15* *2.07*	*0.97*
Route 2	*1.62* *2.47*	*0.82*
Route 3	*2.11*	*0.92* *1.23*
Current	----	*1.38* *1.35*

TABLE 13

PROPERTIES OF TG3 ROLLED JOINTS

Tube Type	Leak Rate (cc/s He)		Tensile Strength (lb)	Yield or Failure	Maximum Tensile Residual Stress (psi)			
					Inside		Outside	
	As Rolled	After Thermal Cycling			Axial	Circumf.	Axial	Circumf.
Route 1	Below Spec.*	2.7×10^{-8}	140,697	Yield	13,000	1,500	42,000	2,000
	Below Spec.*	Below Spec.	151,704	Failure	14,000	1,000	39,500	3,500
	Below Spec.*	1.24×10^{-6}	143,999	Yield	7,000	-1,000	33,500	1,000
	Below Spec.*	8.64×10^{-7}	151,887	Failure				
Route 2	Below Spec.	Below Spec.	146,751	Yield	17,000	5,500	31,000	2,500
	Below Spec.	Below Spec.	155,923	Failure	5,500	-2,000	39,000	1,000
	Below Spec.	Below Spec.	143,632	Failure	6,500	-1,000	38,500	0
	Below Spec.	Below Spec.	145,833	Failure				
Route 3	Below Spec.	Below Spec.	147,668	Failure	9,000	1,000	43,500	1,000
	Below Spec.	Below Spec.	150,420	Yield	15,000	7,000	32,500	5,000
	Below Spec.	Below Spec.	148,402	Yield	19,000	11,000	39,000	5,500
	Below Spec.	Below Spec.	153,538	Failure				

[* Specification Leak Rate's 2.0×10^{-8} cc/s He.]

TABLE 14

MEASUREMENTS OF GRAIN WIDTH AND ASPECT RATIO (LENGTH/WIDTH) IN THE TUBES

Route	*Alpha Grain Width (μm)*	*Aspect Ratio (l/w)*
1	*0.25*	*15-20*
2	*0.34*	*5-10*
3	*0.37*	*5-10*
Current	*0.34*	*15-20*

TABLE 15

MEASUREMENTS OF DISLOCATION DENSITY (m^{-2}) ON TUBES

	Front	*Back*
Route 1	2.6×10^{14}	3.0×10^{14}
Route 2	6.0×10^{14}	5.4×10^{14}
Route 3	3.3×10^{14}	3.2×10^{14}
Current	$5.0 - 7.0 \times 10^{14}$	$5.0 - 7.0 \times 10^{14}$

TABLE 16

TEXTURES OF TUBES IN TERMS OF FRACTION OF BASAL POLES IN PRINCIPAL TUBE DIRECTIONS

	Front			Back			
	f_R	f_T	f_L	f_R	f_T	f_L	f_R/f_T (Average)
Route 1	0.39	0.58	0.04	0.40	0.57	0.03	0.7
Route 2	0.35	0.60	0.04	0.39	0.57	0.04	0.6
Route 3	0.36	0.60	0.04	0.36	0.60	0.03	0.6
Current	0.30	0.63	0.08	0.34	0.60	0.05	0.5

TABLE 17

INFLUENCE OF ANNEALING TEMPERATURE ON TEXTURE OF COLD WORKED ROUTE 2 TUBE

Material Condition	f_R	f_T	f_L	f_R/f_T
20% Cold Work	0.36	0.59	0.05	0.6
20% cw + 650°C-0.5 h	0.32	0.64	0.04	0.5
20% cw + 700°C-0.5 h	0.27	0.70	0.03	0.4

TABLE 18

RELAXATION RATE OF BENT BEAM SPECIMENS FROM IN REACTOR TESTS AT ~530K TO A FLUENCE OF 5.3×10^{24} n-m^{-2}

	Rate (h^{-1})*	
	Transverse	Longitudinal
Route 1	$1.55 \pm 0.24 \times 10^{-4}$	$2.74 \pm 0.31 \times 10^{-4}$
Route 2	$1.54 \pm 0.25 \times 10^{-4}$	$2.47 \pm 0.32 \times 10^{-4}$
Route 3	$1.54 \pm 0.16 \times 10^{-4}$	$2.99 \pm 0.11 \times 10^{-4}$
Current	$1.74 \pm 0.18 \times 10^{-4}$	$2.89 \pm 0.15 \times 10^{-4}$

* ± 95% Confidence Limit

TABLE 19

SUMMARY OF THE DIFFERENCES IN TEXTURE, GRAIN SHAPE AND DISLOCATION DENSITY OF THE TUBES

	Current Tubes	Route 1	Route 2	Route 3
Crystallographic Texture f_R / f_T	0.52	0.67	0.58	0.60
Aspect Ratio of The Alpha Grains	15>20	15>20	5 - 10	5 - 10
Relative Dislocation Density	1.0	0.3	1.0	0.5